

Electrical Properties of Electrodeposited ZnCuTe Ternary Nanowires Embedded in Polycarbonate Membrane

Regular Paper

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Abstract The electrical properties of electrodeposited zinc-copper-telluride (ZnCuTe) nanowires embedded in polycarbonate track-etch membrane (Whatman) having pore-diameters of 200, 100 and 50nm have been studied and reported in this paper. Scanning electron microscopy (SEM) confirmed the formation of the nanowires of uniform diameter equal to the diameter of the templates used. *I-V* measurements at room temperature (303K) reveal that the nanowires of diameter 200, 100 and 50nm have linear and ohmic characteristics. Higher electron transport was observed in larger diameter nanowires compared to smaller ones which may be attributing to size effect. The temperature-dependent electrical transport measurements over a temperature range 308-423K reveal that the temperature dependence of electrical conductivity increases with increasing temperature and decreases as the nanowires size decreases. The study reveals that ZnCuTe nanowires have negative temperature coefficient of resistance (TCR).

Keywords Electrodeposition; Ternary Nanowires; SEM; Electrical Transport Characteristics

1. Introduction

In recent years, nanostructured materials with least one dimension in the range of 1–100 nm have gradually become one of the hottest topics in the field of materials science, because of the fact that they can exhibit unique electrical, optical, magnetic, thermal, mechanical, and chemical properties compared with their bulk parent counterparts [1]. One dimensional nanomaterials, mainly nanowires, nanotubes, nanorods, and nanobelts have attracted special attention because they do not only possess unique structural one dimensionality and possible quantum confinement effects in two dimensions, but also they are expected to play an important role as building blocks for the future nanoscale devices [2-6]. The electrical transport properties of nanomaterials are important in order to understand their possibility for their use in nanoscale electronic devices.

The electrical properties of II-VI compound semiconductor nanostructures such as ZnTe [7-9], ZnS [10], CdS [9, 11-12], and CdTe [9-10, 13] have been studied widely and have established their applications in nanoscale optoelectronics and photovoltaic technology. Their properties make them

an important class of materials and competing candidates for silicon and other semiconductor materials in photovoltaic and optoelectronic applications. CdS and CdTe are well studied and established materials for CdS/CdTe photovoltaic technology [10-11, 13]. Copper based chalcogenides p-type semiconductors such as CuTe & ZnCuTe have potential applications in solar cells, photodetectors, electroconductive electrodes and other advanced optical applications [14-18]. The copper telluride and zinc copper telluride are widely used as back-contact materials to thin film CdS/CdTe photovoltaic devices for achieving higher efficiency and stability of the device [19-20]. The electrical properties of copper chalcogenides with various metallic electrodes have been investigated by many researchers [7, 17, 21]. ZnCuTe is a promising material for bifacial characterization as well as tandem cell interconnects and more stable low resistance Cu-doped contacts in CdS/CdTe solar cell [22-23]. Gessert et al. [24] and Tang et al. [25] have reported the properties of p-ZnCuTe back-contact material for making ohmic contacts to CdTe base in CdS/CdTe thin film solar cells. The research on efficiency and stability of photovoltaic devices is a field of current research in photovoltaic engineering. Nanowires solar cells will give much higher efficiency and stability compared to present thin film solar cell. The higher efficiency can be achieved by multi-junction nanowires based solar cell [26]. Haverkort et al. [27] claimed to develop nanowires solar cell with efficiency more than 65% with 5 to 10 junctions.

Using template-assisted one-step dc electrodeposition, ternary semiconductor ZnCuTe nanowires have been synthesized within the pores of polycarbonate track-etch membranes at room temperature (303K). The template-assisted electrodeposition has been demonstrated to be a simple and versatile approach for the preparation of the 1D nanostructure of numerous materials, including metals [28], semiconductors [29] and conductive polymers [30]. It has been considered as one of the most efficient methods in controlling the growth of nanowires because it takes place exclusively in the normal direction of the substrate. Compared to conventional methods, the higher degree of reproducibility makes template-assisted methods the most attractive way to fabricate nanowires. Through the use of template such as polycarbonate track-etch membrane or anodic alumina oxide (AAO), the reproduction of the structure with the best possible reproducibility can be achieved [31]. The template methods are generally inexpensive, allowing deposition of a wide range of materials, and presenting the ability to create very thin wires down from 10 μ m to 5nm with higher aspect ratios [31-32] as compared to other methods e.g., chemical vapor deposition, pulse laser deposition, sol-gel, hydrothermal and molecular beam epitaxy. The template method for synthesis of nanowires provides an effective control over the uniformity, dimensions and shape of the nanowires.

On the other hand, electrodeposition is one of the most widely used methods to fill the nanoporous template with conducting and semiconducting materials to form continuous nanowires. This pattern of electrodeposition is a promising technique for fabricating nanostructures in a "Bottom up" approach [33].

2. Experimental techniques

Zinc copper telluride (ZnCuTe) ternary compound semiconductor nanowires of diameter 200, 100 and 50nm have been deposited on copper substrates from an aqueous electrolyte solution of Zn, Cu and Te. The aqueous electrolyte solution was prepared from 0.2M ZnSO₄·7H₂O (AR, Merck), 0.15M CuSO₄·5H₂O (AR, Merck) and 1mM TeO₂ (99%+, Sigma-Aldrich) dissolved in 3-4 drops of H₂SO₄ (AR, Merck) in 50ml pure double distilled water. The pH of the electrolyte was adjusted between 2 to 3 by using H₂SO₄ and was measured 2.1 from digital pH meter in all the experiments. The as-received self-adhesive copper tape (3M 1181) without any further cleaning was used as substrates. The copper tape was fixed on the aluminium base of the self-made electrochemical cell that acts as cathode. The polycarbonate track-etch membranes (Whatman) were used as overlays fixed gently on the copper substrate to select an area of approximately 1cm² on the copper substrate via O-ring fitted in the top portion of the cell. In this way, an area of approximately 1cm² on the copper substrate is allowed for exposure with the electrolyte through nanoporous membrane. A platinum wire was used as an anode. The electrodeposition was carried out for 15 min at 0.71V dc voltage at room temperature and a current density in the range of 7-9mA/cm² was observed during the experiment at room temperature (303K). The nanowires got deposited in the nanopores of the membrane. The samples were removed from the cell carefully after the deposition and washed with distilled water. The samples in membranes were then dried in open air for about half an hour and used for electrical characterization. The nanowires were retrieved by dissolving polycarbonate membranes in dichloromethane (CH₂Cl₂), rinsed with double distilled water and dried at room temperature. The bare nanowires on copper substrates were used for SEM, EDX and XRD characterization.

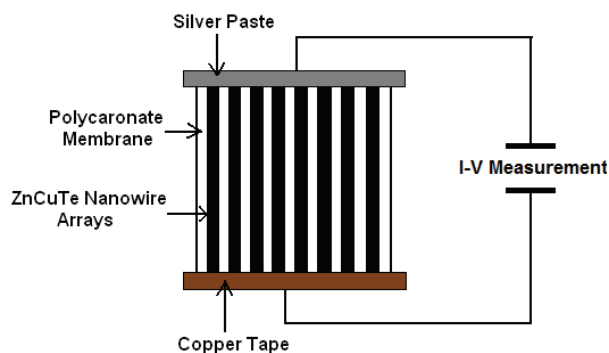


Figure 1. Schematic of the two-probe dc technique used for the measurement of electrical transport properties of ZnCuTe nanowires

The JEOL JSM-6100 and Zeiss EVO 40 analytical scanning electron microscopes (SEM) were used to view the morphologies of as-prepared ZnCuTe nanowires on copper substrate. The structural properties were studied by X'PERT-PRO Phillips X-ray Diffractometer using Cu-K α radiation at 45 mA, 45 keV with a step size of 0.0170 using wavelength (K α) 1.5406Å in the range of scanning angle 20–70 degree. The two-point probe method was used to study the electron transport properties of as-prepared ZnCuTe nanowires embedded in the membrane itself. A self-made two probe apparatus was used to study the electrical properties. Figure 1 shows the schematic of two-probe dc technique for the measurements of electrical transport properties of ZnCuTe nanowires. The apparatus was consisting of two electrodes with a heater in a cylindrical aluminium body for measuring temperature dependent electrical conductivity. The contact to ZnCuTe nanowires was made using conducting silver paste on an area approximately 0.5mm x 0.5mm. The ZnCuTe nanowires were therefore sandwiched between the two metal plates i.e. Cu-ZnCuTe-Ag, thus formed a metal-semiconductor-metal system. The nanowires embedded in polycarbonate membrane were suspended between the two electrodes for electrical measurement in two-point probe apparatus. *I-V* measurements were recorded at room temperature (303K) at different voltages across the nanowires. The temperature dependent electrical conductivity was measured by increasing the temperature surrounding the nanowires in the temperature range 308–423K. The applied dc voltage of 90mV was kept constant across the nanowires during temperature -dependent electrical measurements. The programmable voltage source and electrometer (Keithley, Model 617) was used for all measurements.

3. Results and discussion

3.1 SEM, XRD & EDX characterization

Figure 2(a), 2(b) and 2(c) shows the micrographs of the as-prepared ZnCuTe nanowires of diameter 200, 100 and 50nm, respectively. The SEM micrographs of as-prepared ZnCuTe nanowires in figures 2(a & b) and 2(c) were taken from JEOL JSM-6100 and Zeiss EVO 40 analytical scanning electron microscope (SEM) respectively. The SEM micrographs show that the nanowires are homogeneous, vertical, free standing and highly ordered. The growth of the nanowires is normal to the substrate. The nanowires are found to have equal diameter to that of the templates used and are found to be of very high aspect ratio.

The figures 3(a), 3(b) and 3(c) shows the XRD pattern of as-prepared ZnCuTe nanowires of diameter 50, 100 and 200 nm respectively on copper substrate. The peaks of ZnCuTe indexed as (006), (106), (108), (119), (209), (326)

and (331) are observed in the XRD patterns of diameters 200, 100 and 50nm that matches with the standard ICDD zinc copper telluride (ZnCuTe) data file (JCPDS Card Number: 45-1301) which confirm the formation of ZnCuTe nanowires with hexagonal structure. The various strong peaks of copper (JCPDS Card Number: 04-0836, 02-1225) are also observed in the XRD pattern that may arises from the copper substrate. The broadened peaks are observed in the XRD spectrum with decrease in the diameter size of ZnCuTe nanowires which attribute to the size effect. The strong reflection peaks suggest that the well-crystallized ZnCuTe nanowires were successfully obtained through the present fabrication method.

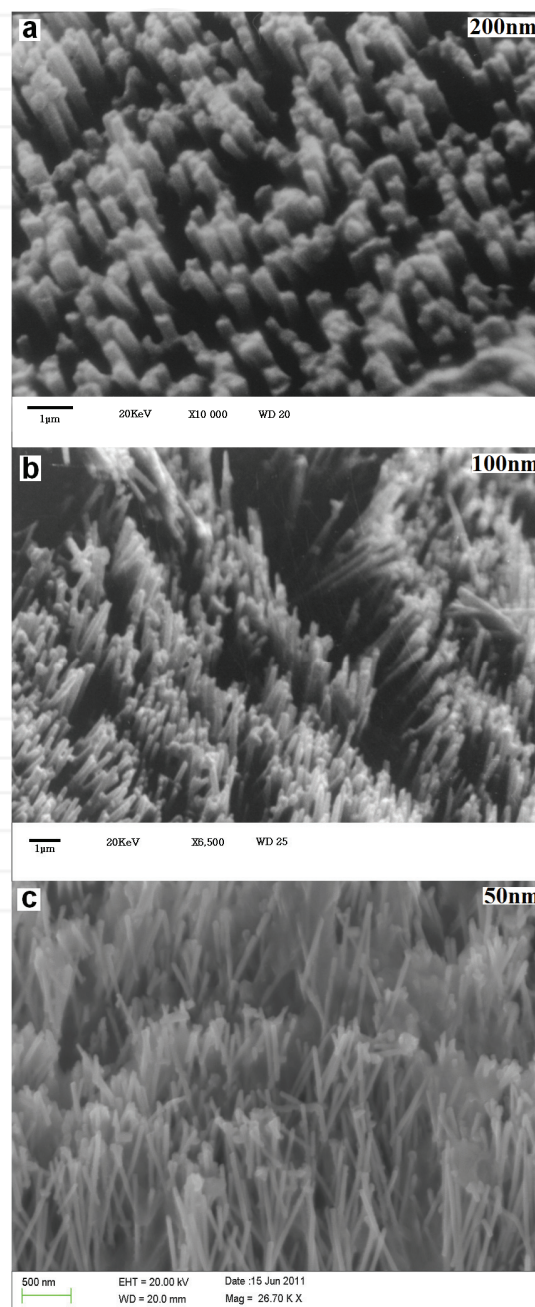


Figure 2. SEM micrographs of ZnCuTe nanowires of diameters (a) 200nm (b) 100nm and (C) 50nm on copper substrates

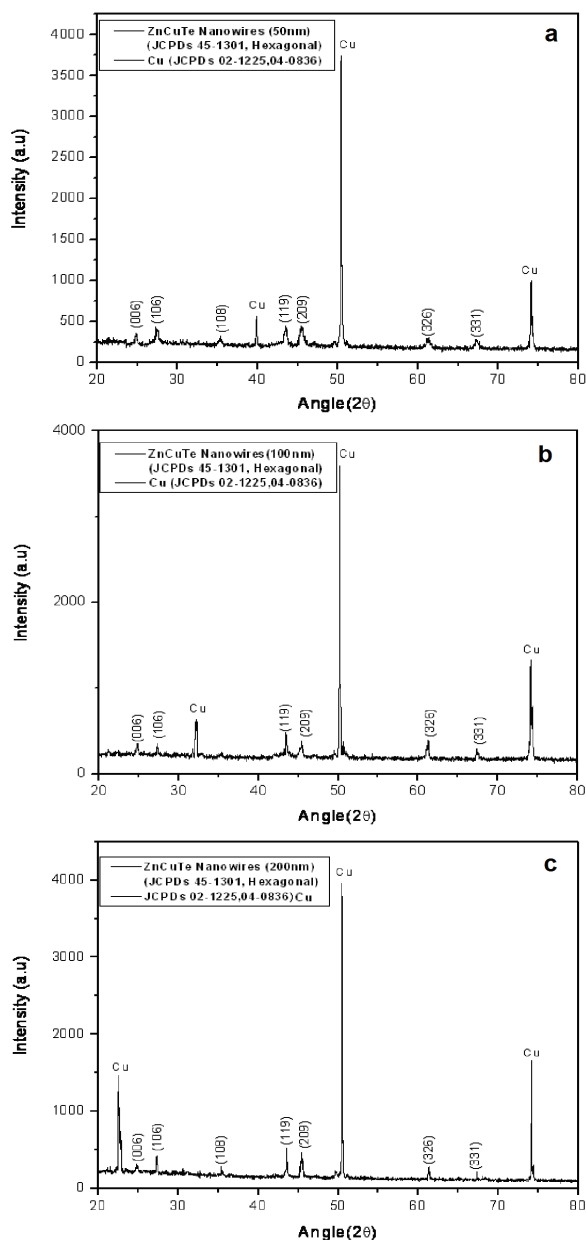


Figure 3. XRD spectrum of ZnCuTe nanowires of diameters (a) 50nm (b) 100nm and (C) 200nm on copper substrates

The EDX analysis of ZnCuTe nanowires (figure 4) revealed that the nanowires are chemically composed of Zn, Cu and Te [34]. The EDX spectrum also shows the signals of Sn and O that come from the substrate. The C signal originates from the carbon tape taken as a support of sample during EDX investigation. The chemical composition of the electrolyte for the all the ZnCuTe nanowires of diameters 200, 100 and 50 nm was kept same during the synthesis of nanowires.

3.2 Electrical studies of zinc copper telluride nanowires

The electrical properties of zinc copper telluride (ZnCuTe) ternary nanowires have been studied at room temperature using two-point probe method. The

temperature dependence of electrical conductivity was measured over a temperature range of 308-423K. Both the studies were carried out in a self-made two electrode apparatus made of aluminium.

3.2.1 Current-voltage (*I-V*) characteristics

The room temperature (303K) *I-V* measurements of as-prepared zinc copper telluride (ZnCuTe) nanowires embedded in polycarbonate membranes were recorded using dc two-point probe method. Figure 5 shows the linear and symmetrical *I-V* relationship of as-prepared zinc copper telluride nanowires. *I-V* curves have shown linear relationship of current and voltage which suggests that the ZnCuTe nanowires have ohmic character. Thus, the Cu-ZnCuTe-Ag system can be modeled as having back-to-back ohmic contacts. The study further suggests that the zinc copper telluride nanowires can be used as ohmic contacts in future nanodevices and nanowires based photovoltaic devices.

The figure 5 clearly shows that the electrical conductivity in 50nm nanowires is lower than 100nm nanowires and that of 100nm is lower than 200nm nanowires. This dependence of electrical conductivity on size may be accounted for the size effect in nanomaterials. The material properties changes drastically when size of the material is reduced to nanometer range. The electron-electron interaction contributes mainly to the electron transport through the nanowires at room temperature. The electron scattering at the surface and grain boundaries dominates in nanomaterials due to the size effect greatly affects electron transport. As the size is reduced, electron scattering at the surface or grain boundaries increases due to the presence of more unbonded atoms at the surface of the nanowires. These unbonded atoms produce defects within the nanowires, and may cause the nanowires to transport electrons more poorly than that in the bulk material. Mayadas & Shatzkes [35] and Fuchs-Sondheimer [36, 37] suggested two theories of electrical conduction in materials when their size decreases to nanometer range. Mayadas and Shatzkes suggested that the reflected electrons at the grain boundaries reduce the electrical conduction due to grain boundary scattering that results higher resistivity in nanomaterials. The Fuchs-Sondheimer theory suggested that the mean free path of electrons is reduced due to electron surface scattering on the wire surface. This scattering of electrons from the surface increases the resistivity in nanomaterials and hence limits the electron transport through nanomaterials. Thus, both the theories explain that the electrical conduction decreases due to increasing resistivity with decrease in size of the nanowires. Therefore, as a result of surface or grain boundary scattering in nanowires, the wire resistivity increases which in turns decrease the electron transport through the nanowires.

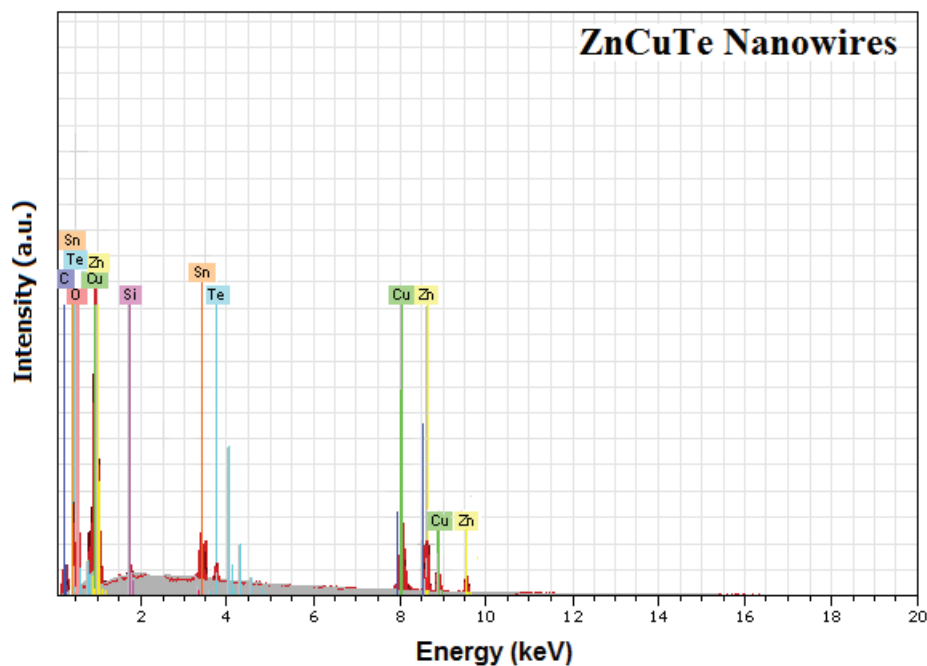


Figure 4. Energy dispersive X-ray (EDX) spectrum of zinc copper telluride (ZnCuTe) nanowires

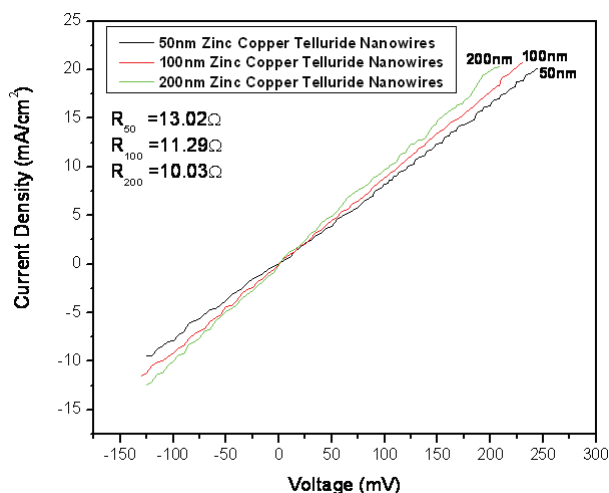


Figure 5. Current-voltage (*I-V*) characteristics of 200, 100 and 50nm zinc copper telluride nanowires at room temperature

The transport of the electron through the nanowires is also influenced due to quantization of energy in the nanowires. The energy of the electrons through nanowires assumes discrete values due to quantum size effects which become more prominent when the size of the material approaches in nanometer regime. When a charge carrier is confined, then its energy is quantized [38]. The electrical conduction then becomes the sum of the transport by separate channels of different quantized energy levels. As the size of the nanowires decrease, the number of channels available to the transport of electrons decreases. Therefore, the overall effect is the decrease in electron transport through the nanowires with decrease in the size of the nanowires.

The average resistance from the *I-V* graph of zinc copper telluride nanowires was calculated to be 13.02, 11.29 and 10.03 Ω for 200, 100 and 50nm respectively.

3.3.2. Temperature-dependent electrical resistance

The measured temperature dependence of resistance over a temperature range of 308-423K for 200, 100 and 50nm diameters of zinc copper telluride nanowires is shown in figure 6. The measurement results reveal that the resistance (R_T) of nanowires of all diameter rises as the temperature decreases. The increase of resistance in the nanowires at low temperature attributes to the reduction of free carriers in the conduction band [39], where only carriers with enough energy can stay in the conduction band for electrical conduction. The drop in carrier density at low temperatures suggests the overlapping of the conduction band and valence band where thermal energy is the main factor to determine the free carrier density. Therefore, the temperature-dependent resistance measurement shows that the electrical conductivity of the ZnCuTe nanowires is carrier density dependent. The exponential decrease in carrier density in low temperature range suggests that the ZnCuTe nanowires have semiconducting behaviour. It is clear from figure 6 that the resistance (R_T) of ZnCuTe nanowires decreases with increasing temperature and exhibits an obvious negative temperature coefficient of resistance (TCR). It is also observed from the figure 6 that the value of resistance (R_T) increases with decrease in the size of ZnCuTe nanowires.

The temperature dependent resistivity of the ZnCuTe nanowires was computed based on the measurement data using equation (1):

$$\rho = R \frac{A}{l} \quad , \quad (1)$$

where l is the length, A is the cross-sectional area and R is the measured resistance value. The temperature dependent resistivity values lie in the range of 4.78×10^{-1} - 4.63×10^{-1} , 4.86×10^{-1} - 4.69×10^{-1} and 5.02×10^{-1} , and 4.74×10^{-1} ohm-meter in the temperature range of 308-423K respectively.

The electrical conductivity of as-prepared 200, 100 and 50nm diameter ZnCuTe nanowires as a function of temperature can be deduced from resistance measurements and is plotted in the Figure 7. The electrical conductivity reaches to its peak value of 0.334, 0.331 and 0.324 $\Omega^{-1}\text{m}^{-1}$ for 200, 100 and 50nm diameter nanowires at around 393, 408 and 423K respectively. This decrease in the peak value is due to decrease in the size of the nanowires which attributes to size effect. Therefore, it is observed from the graph that the temperature-dependent electrical conductivity decreases with decrease in size of ZnCuTe nanowires. It is also observed that the electrical conductivity increases with increase in temperature in all nanowires and this increase may be due to the increased electron-phonon interaction. Both the electron-electron and the electron-phonon interactions take part in the electron transport through the nanowires. As the temperature is increased, phonon induced effects dominates and electron-phonons interaction becomes more prominent. Further, such interactions dramatically enhances the electron transport in addition to phonon through nanowires due large carrier generation when temperature is increased. The activation energy (E_a) in ZnCuTe nanowires was calculated, using Arrhenius equation (1) for the conductivity σ in semiconductors [40]:

$$\sigma = \sigma_0 \exp(-E_a / kT) \quad , \quad (2)$$

where σ is electrical conductivity in $\Omega^{-1}\text{m}^{-1}$, σ_0 is a pre-exponential factor, E_a is the activation energy in eV, k is Boltzmann constant, and T is temperature in Kelvin. From equation (1), the calculated activation energy (E_{a1}) are 0.59, 0.66 and 0.73meV over a temperature range of 340-423K in 200, 100 and 50nm diameter zinc copper telluride nanowires, respectively. The slope significantly changes in low temperature range below approximately 340K and the measured activation energy (E_{a2}) values in low temperature range of 308-340K are 5.32, 8.15 and 11.70 meV in 200, 100 and 50nm diameter ZnCuTe nanowires, respectively.

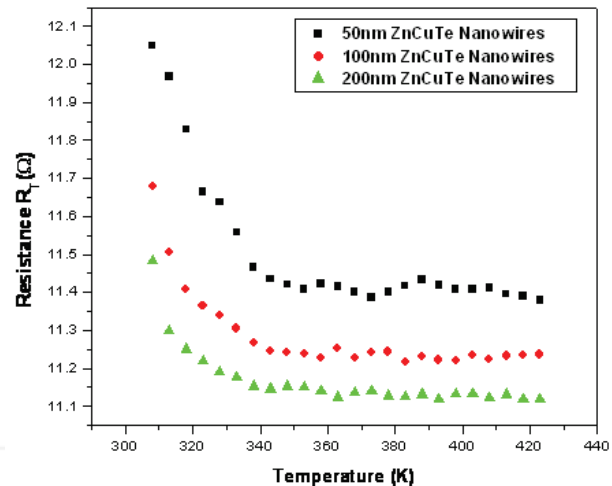


Figure 6. Temperature dependence of resistance of ZnCuTe nanowires of diameters 200, 100 and 50nm

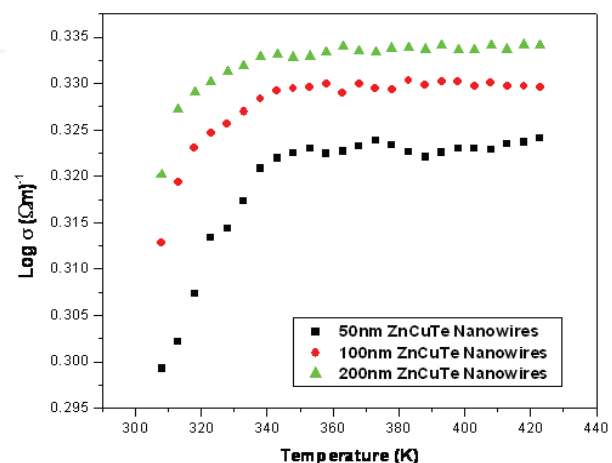


Figure 7. Temperature dependence of electrical conductivity of ZnCuTe nanowires of diameters 200, 100 and 50nm

4. Conclusion

The room temperature and temperature-dependent electrical properties of ZnCuTe nanowires of diameter size 200, 100 and 50nm synthesized electrochemically via template synthesis using polycarbonate track-etch membrane have been studied and reported. The SEM studies have confirmed that the ZnCuTe nanowires are ordered, vertically aligned and of high aspect ratio. The diameters of synthesized nanowires have diameter equal to the diameter of the pore-templates used. I - V measurements of ZnCuTe nanowires embedded in polycarbonate membrane have shown linear, symmetric and ohmic characteristics. These nanowires could find applications as ohmic contacts in future nanowires based photovoltaic devices. The electron transport through the nanowires decreases with decrease in size of the nanowires due to quantum size effect. The electrical studies reveal that the electrical conduction in all the nanowires are temperature dependent and increases with

temperature. The resistance of ZnCuTe nanowires decreases with increasing temperature and thus have negative temperature coefficient of resistance (TCR).

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6. References

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